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14. ABSTRACT

Under STIR award (62022CHII), we achieved the first experimental observation of the dependence of strong field ionization rate on the sign of the magnetic quantum number. We measure the strong field sequential double ionization yield of argon by two time-delayed near-circularly polarized laser pulses. It is found that double-ionization yield is enhanced more than three times if two lasers have the opposite helicity. Analysis shows that the single ionization of both the neutral and ion prefer the same sign of the magnetic quantum number.

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Report Title

Strong Field Ionization Rate Depends on the Sign of the Magnetic Quantum Number

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Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received		Paper
03/01/2013	1.00	Thushani Herath, Lu Yan, Suk Kyoung Lee, Wen Li. Strong-Field Ionization Rate Depends on the Sign of the Magnetic Quantum Number, Physical Review Letters, (07 2012): 0. doi: 10.1103/PhysRevLett.109.043004
03/01/2013	2.00	Suk Kyoung Lee, Arthur G. Suits, H. Bernhard Schlegel, Wen Li. A Reaction Accelerator: Mid-infrared Strong Field Dissociation Yields Mode-Selective Chemistry, The Journal of Physical Chemistry Letters, (09 2012): 0. doi: 10.1021/jz301038b

TOTAL: 2

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Number of Presentations: 0.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Paper

TOTAL:

Patents Submitted

Patents Awarded

Awards

Wen Li 2013 Sloan research fellowship

Graduate Students

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
Suk Kyoung Lee	0.75
FTE Equivalent:	**0.75**
Total Number:	**1**

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Wen Li	0.08	
FTE Equivalent:	**0.08**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period:	0.00
The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:......	0.00
The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:......	0.00
Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):......	0.00
Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:......	0.00
The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense	0.00
The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:	0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME PERCENT_SUPPORTED

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

Technology Transfer

Strong Field Ionization Rate Depends on the Sign of the Magnetic Quantum Number

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Abstract

Under STIR award (62022CHII), we achieved the first experimental observation of the dependence of strong field ionization rate on the sign of the magnetic quantum number. We measure the strong field sequential double ionization yield of argon by two time-delayed near-circularly polarized laser pulses. It is found that double-ionization yield is enhanced more than three times if two lasers have the opposite helicity. Analysis shows that the single ionization of both the neutral and ion prefer the same sign of the magnetic quantum number. Furthermore, the intensity dependence of this sensitivity has been measured in both xenon and krypton. It was found that spin-orbital coupling does not suppress the dependency of strong field ionization on atomic orientation. These results are reported in two research paper, one was published in Physical Review Letters (109,043004, 2012) and the other manuscript is in preparation.

In an intense laser field, electrons in atoms/molecules interact strongly with the field and are instantaneously ionized from a suppressed Coulomb barrier[1]. The theory of tunneling ionization[2-4] predicts a strong dependence of the ionization rate on the alignment of the electronic orbitals (described by the distribution of the absolute value of the magnetic quantum number, $|m|$). Previously, this sensitivity have been utilized to produce noble gas cations with non-statistical $|m|$ distribution and then such alignment was probed by absorption spectroscopy[5-7]. Van der Hart[8] invoked this $|m|$ dependence of the ionization rate to interpret the double photo-detachment rate in negative ions. Recently, we have demonstrated this by measuring the angular dependent ionization rate of aligned sulfur atoms produced from photodissociation of carbonyl sulfide and ethylene sulfide[9]. The sensitivity of strong field ionization (SFI) to atomic orbital orientation or helicity, *i. e.* the sign of magnetic quantum number, is less obvious. The first and only theoretical paper that discussed the sensitivity of SFI to atomic orientation was published recently by Barth *et al.*[10]. They predicted the strong field ionization rate of an m=-1 orbital in right circularly polarized light field is three times higher than that of m=1. On the other hand, it is well known that circularly polarized light does preferentially ionize co-rotating electrons (*e. g.* positive m for right circularly polarized light) in one-photon ionization and field ionization of Rydberg states[11, 12]. No experimental result has been reported on the m sign dependence of SFI thus far. However, this subject is fundamentally important to the field of attosecond dynamics because circularly polarized light is widely used in the production of isolated attosecond pulses[13, 14] and in the measurement of correlated electron dynamics by the angular streaking technique[15, 16]. In the theoretical modeling of these experiments, the sign of the magnetic quantum number was generally not taken into consideration. It is worth pointing out that any

m dependence will vanish in an ultra-strong laser field (10^{16}-10^{21} W/cm^2), due to *m* level scrambling by the laser field[17].

Our interest in the sensitivity of SFI on atomic orbital orientation is due to its potential application as an ultrafast probe of chemical dynamics. Oftentimes, photodissociation of molecules produce aligned/orientated atomic fragments that reflect the details of the dynamics such as state symmetry, curve crossing and coherence[18-28]. Conventionally atomic alignment/orientation are measured by resonance enhanced multi-photon ionization (REMPI) in the asymptotic region without time resolution[21, 29] (also see a comprehensive review[30]). Strong field ionization can probe the electron density rearrangement in real-time during a chemical reaction[31]. It would provide even more information about the time dependent electronic wave functions if it can also track the time-resolved *m* distribution (atomic alignment/orientation).

During the grant period, we discovered for the first time that SFI rate by circularly polarized light depends on the sign of the magnetic quantum number. We achieve this by comparing the sequential double ionization (SDI) yields of argon by two nearly-circularly polarized laser pulses with same helicities and opposite helicities. If SFI prefers one sign of the magnetic quantum number to the other, the first pump pulse would produce single ions with nonstatistical *m* distribution (the ion's orbital angular momentum is orientated). The probe pulse will see this *m* distribution and thus the total ion yield will be different depending whether the pump and probe have the same or opposite helicities. Our data show that the ion yield with opposite helicities is three times higher than that with the same helicities. Further data analysis shows that the ionization rate of one sign of the magnetic quantum number is at least 4 times higher than that of the opposite sign. Our result is the first experimental observation of this dependence. We also showed recently that the strong spin-orbital coupling in krypton and xenon does not suppress the dependency of strong field ionization on atomic orientation.

The experiment was carried out in our newly built velocity mapping coincidence apparatus. The laser was a 4mJ/pulse, ~70 fs, one kHz Ti:Sapphire amplification system (KMLabs, Red Dragon). The laser beam was split into one pump beam and one probe beam with a Mach-Zenhnder type interferometer. The pump beam was bounced off two turning mirrors, which were both mounted on a motorized translation stage and thus the time delay between the pump and probe beam could be varied continuously. Both beams were focused onto the atomic beam by two plano-convex lenses (f/40 for pump and f/50 for probe). The power of the pump and probe beam are 300 μJ and 600 μJ, respectively, corresponding to laser intensities of ~9×10^{13} W/cm^2 and ~1.4×10^{14} W/cm^2. Before the lenses, we inserted a quarter wave plate in each beam to produce circularly polarized light. The measured ellipticity of the probe beam is 0.8. The helicity of the pump beam was changed by rotating the quarter wave plate by 90 degrees. The measured ellipticities of the pump beam with right and left circularly polarization are 0.88 and -0.80, respectively. The produced argon dications were extracted by the multi-lens velocity mapping electrode assembly and impacted upon a micro-channel plate (MCP)/phosphor detector. A typical time dependent trace of argon dication yield is shown in figure 1(a). We collected the ion yield under four laser conditions: both

lasers on (ion yield labeled as I_{pp}), pump beam off (I_{pr}), probe beam off (I_{pu}) and both beam off (I_{d}, dark counts). The argon dications produced sequentially by the pump and the probe were calculated using $I_{SDI}=I_{pp}-I_{pr}-I_{pu}+I_{d}$. The averaged data is shown in figure 1(b). The calculated I_{SDI-LR} (LR stands for the opposite helicity between the pump and the probe) and I_{SDI-RR} are 4.21±0.98 and 1.16±0.92, respectively. The difference in total dication yields is obvious. An intuitive way to understand this result is as follows: the pump laser first preferably depletes the sublevel of one sign of the magnetic quantum number in the single ions; then, the probe laser with an opposite helicity can further ionize the sublevel with the opposite sign of *m*, while the probe laser with the same helicity sees a depleted population of the sublevel with the same sign of *m* and thus the ionization yield of the dication is lower.

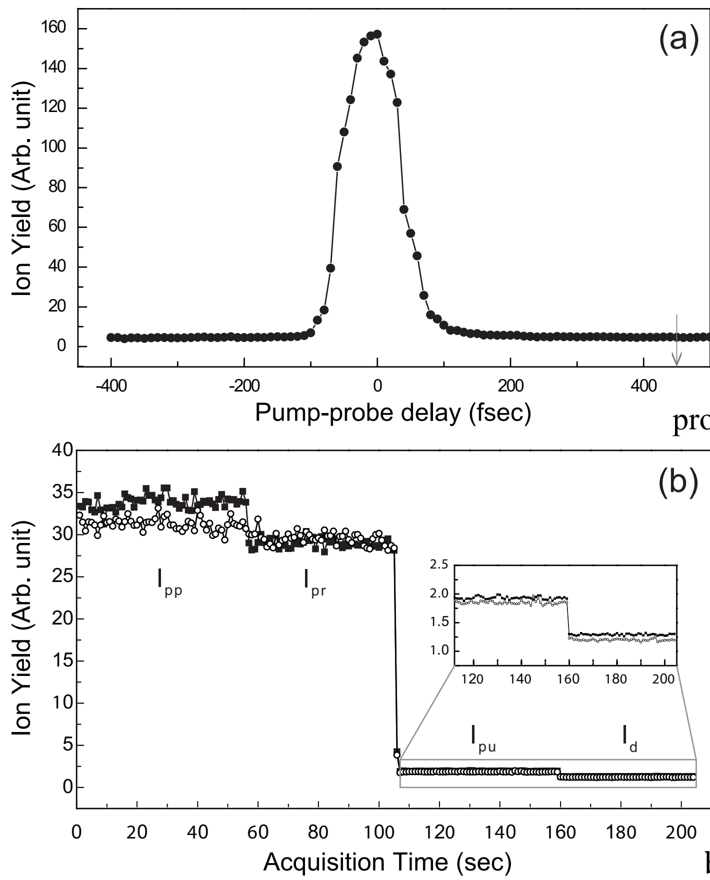

Figure 1. (a) The time dependent Ar^{2+} yield. The grey arrow marks the time delay used for the data extraction. (b) The ion yield of Ar^{2+} under different laser conditions: I_{pp} with both laser on; I_{pr}, the pump laser off; I_{pu}, the probe laser off; I_{d}, both laser off. Solid squares: opposite pump-probe helicity; hollow circles: same pump-probe helicity. The inset is a zoom-in of the ion yields with the probe laser blocked and both lasers blocked, showing the very small variation in ion yield for the pump alone case.

To further analyze the data and extract the ionization rate ratios between the sublevel of *m*=-1 and *m*=1 in circularly polarized laser field, we need to establish the relation between the ionization rates and the measured ion yields of the dications. For argon, the electrons are ionized out of the 3*p* subshell, with three magnetic quantum numbers: -1, 0, +1. If we label their ionization rates in right circularly polarized light as w_{-1R}, w_{0R} and w_{+1R}, respectively, the following relations are true: $w_{-1R} = w_{+1L}$; $w_{0R} = w_{0L}$; $w_{+1R} = w_{-1L}$ by symmetry. We note that the ionization rates for the pump and the probe are different due to the different ionization potentials and laser intensities. The ratio of Ar^{2+} yield between two helicity configurations thus can be written as:

$\frac{I_{SDI-LR}}{I_{SDI-RR}} = \frac{(\alpha+\beta)\alpha'+(1+\alpha)\beta'+(1+\beta)}{(1+\beta)\alpha'+(1+\alpha)\beta'+(\alpha+\beta)}$ (1), in which the primes denote the ionization rates for the probe ionization and $\alpha = \frac{w_{-1R}}{w_{+1R}}; \alpha' = \frac{w'_{-1R}}{w'_{+1R}}; \beta = \frac{w_{0R}}{w_{+1R}}; \beta' = \frac{w'_{0R}}{w'_{+1R}}$. When deriving equation 1, a flat laser pulse temporal envelope is assumed. If the ionization rate of the sublevel with *m*=0 is much smaller than those with *m*=-1 and *m*=1, β and β' approach zero and equation 1 can be further simplified to $\frac{I_{SDI-LR}}{I_{SDI-RR}} = \frac{\alpha\alpha'+1}{\alpha+\alpha'}$ (2). This simplification can be justified according to the calculation performed by Barth *et al.*, in which the ionization rates of *m*=1 and *m*=-1 were calculated to be orders of magnitude higher than that of *m*=0. It can be seen that if either ratio is 1 (no preference between the signs of the magnetic quantum number), the right hand side of equation 2 becomes unity no matter what the value of the other ratio is and thus no enhancement or suppression will be observed in the measured ion yields. A non-unity ratio between ion yields requires that strong field ionization in circularly polarized light prefers one sign of *m* to the other in both neutral and single ions. This is indeed what we observed in our experiment. We can insert the measured ratio of 3.63 into equation 2 and plot α vs. α', shown in figure 2. Both ratios are higher than 3.63 while the exact values depend on their positions at the hyperbola. These larger than unity ratios indicate that in the processes of ionizing neutrals and single ions by circularly polarized light, the same sign of *m* is preferred.

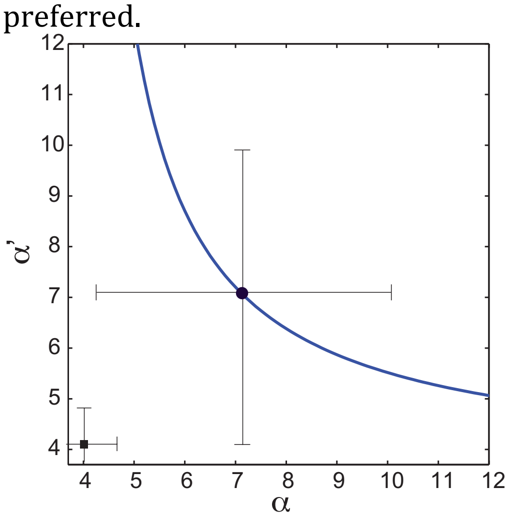

Figure 2. The experimentally constrained ratios of strong field ionization rates between *m*=1 and *m*=-1 sublevels of neutral argon atom (α) and argon single ion (α') by right circularly polarized light. The square marks the ionization rate ratios calculated from the theory by Barth *et al.* (the uncertainty arises from the laser intensity estimate, which is ~20%). The black dot marks the values extracted from the experiment assuming α and α' are equal.

We also developed an intuitive way to help understand the *m* dependence of the strong field ionization rate by circularly polarized light. In this picture, we view the suppressed Coulomb barrier (SCB) as a “doorway” for the electron to “tunnel” under the barrier to the continuum (Figure 3). However, this doorway is not fully open and it is only with a certain probability (*P*) that electrons can tunnel out even though they are spatially close this doorway. For circularly polarized light, this doorway is rotating at the frequency of the laser. For rotating electrons with a nonzero magnetic quantum number, the helicity is determined by the sign of *m*. The relative helicity between electrons and photons affects the encounter frequency between the electrons and the suppressed barrier (γ). The encounter frequency can

be estimated with $\gamma = \nu_e \pm \nu_L$ where ν_e, ν_L are the frequency of the electron rotation and the laser frequency, respectively and the plus sign is used for the opposite helicity. The final ionization probability can be written as γP. From this simple picture, we can qualitatively draw the following conclusions: (1) electrons counter-rotating with the laser helicity will be preferably ionized; (2) The ionization rate increases with the laser frequency for counter rotating but decrease for co-rotating electrons while the ratio between them increases; (3) The ionization rate for m=0 sublevel is greatly reduced due to a destructive interference. These results are in general agreement with the previous theory[10] except that in their calculation the ionization rate of co-rotating electron also increases slowly with the laser frequency. It will be interesting to compare these with experimental data at different laser frequencies or numerical methods.

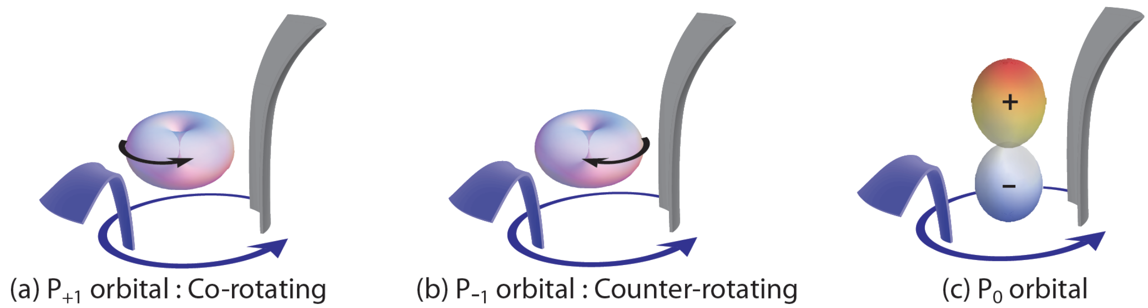

Figure 3. the relative rotation between the electrons and the laser polarization can affect the ionization probability if the suppressed Coulomb barrier is viewed as a "doorway" for tunneling. (a) Co-rotating electrons and the suppressed Coulomb barrier reduce the ionization rate (b) Counter-rotation enhances the ionization rate (c) the opposite phases of the p_z (p_0) orbital lead to a destructive interference and thus a reduced ionization rate.

We extended our measurement to krypton and xenon, two systems with strong spin-orbital coupling. Such coupling generally makes the orbital angular momentum no longer a "good" quantum number and thus might affect the conclusion we drew from argon case, where the spin-orbital coupling is small. Here we took a slightly different measurement. Instead of fixing the ellipticity onto two extremely values (-1 and +1), we scanned the ellipticity continuously and monitored the dication yield at different ellipticities. This allows us to see the difference more clearly. The experiment results are shown in Figure 4. It can be seen that the helicity dependence of strong field ionization are still present in both kypton and xenon. Further data analysis and manuscript preparation is under way.

In summary, by measuring the dication yield with two spatially overlapped but temporally delayed near-circularly polarized lasers, we discover that the strong field ionization rate by circularly polarized light depend on the relative helicity of the photon and the electrons. In the argon case, the measured dication yield with the opposite photon helicity is three times higher than that with the same helicity. From our data analysis we further conclude that the single ionization of both the argon neutral and ion prefer the same sign of the magnetic quantum number. Our experiments on xenon and krypton also showed that such dependence are also present in the systems with large spin-orbital coupling. On the one hand, the sign

dependence will have to be included in the theoretical modeling of many strong field double ionization experiments performed using circularly polarized light. On the other hand, this discovery paves the way for using the strong field ionization by circular fields as an ultrafast probe of photodissociation dynamics that involves orbital orientation.

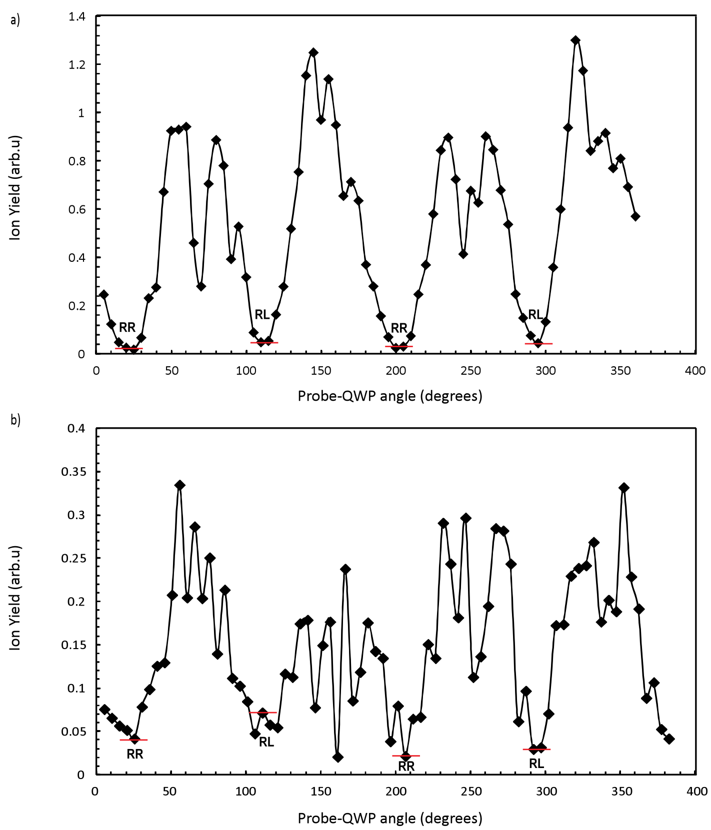

Figure 4. Dication yield difference (I_{pp}- I_{pr}) with varying ellipticities of the probe beam for a) krypton and b) xenon. A minimum SDI yield was obtained at the highest ellipticity as marked in red lines. The opposite helicity (RL) of pump and probe beam gives a higher yield than the same helicity (RR) for both case.

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